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RADIUM-226 IN SNOWSHOE HARES NEAR URANIUM MINE TAILINGS

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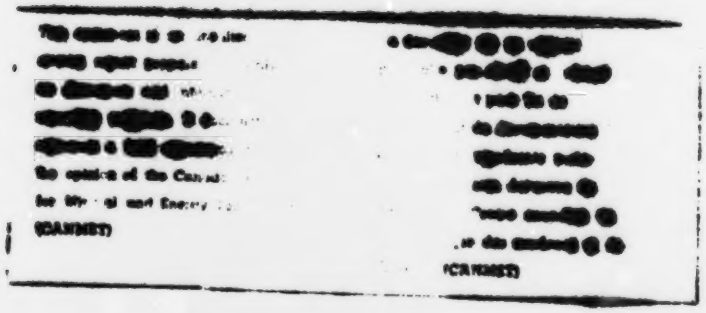

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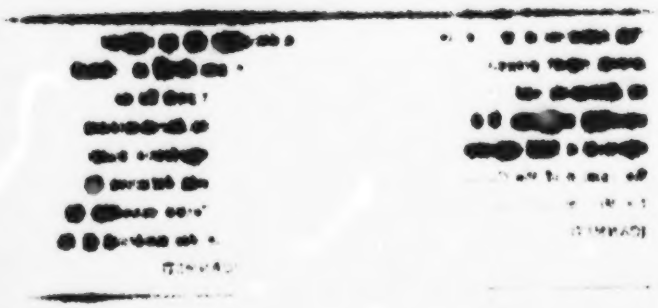

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N.K. Dave+, T.P. Lim++, F.V. Clulow* and N.R. Cloutier**

ABSTRACT

Snowshoe hares (Lepus americanus) trapped near uranium tailings accumulated ^{226}Ra in their bones to levels (175 ± 94 $mBq.g^{-1}$ d.w.) approximately ten times higher than those from areas 3-15 km from the mines: these in turn were at least five times higher than in hares from an area 880 km away. Most stomach contents had levels of ^{226}Ra below detection limits (3.7 $mBq.g^{-1}$ d.w.). This precluded calculation of the transfer coefficient from food to hare. Based on the levels of ^{226}Ra in bones, the maximum internal dose rate to the skeleton and the maximum lifetime dose of hares living near tailings were 3.2×10^{-5} $Gy.d^{-1}$ and 3.5×10^{-2} Gy, respectively. These levels are believed to be too low to pose a health hazard either to the animals or consumers.

Key words: Uranium mill tailings; Ra-226, Snowshow hare; Radionuclide uptake; Low level radioactive waste.

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RADIUM-226 CHEZ LE LIÈVRE D'AMÉRIQUE VIVANT PRÈS DES SITES DE RÉSIDUS DE MINE D'URANIUM

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RÉSUMÉ

Des lièvres d'Amérique (Lepus americanus) capturés au voisinage de sites de résidus d'uranium ont accumulé dans leurs os des concentrations de ^{226}Ra (175 ± 94 $mBq.g^{-1}$ p.s.) environ dix fois plus élevées que les lièvres provenant de régions situées à 3-15 km des mines; ces derniers ont présenté des concentrations au moins cinq fois plus élevées que celles que l'on a enregistrées chez les lièvres provenant de régions situées à 880 km. La plupart des analyses du contenu stomacal ont révélé des concentrations de ^{226}Ra inférieures aux limites de détection (3,7 $mBq.g^{-1}$ p.s.). À cause de cette situation, on n'a pu calculer le coefficient de transfert des aliments au lièvre. À partir des concentrations de ^{226}Ra enregistrées dans les os, on a déterminé que le débit de dose interne maximum pour le squelette ainsi que la dose maximale à vie pour les lièvres vivant à proximité des sites de résidus étaient respectivement de 3,2 x 10^{-5} $Gy.d^{-1}$ et 3,5 x 10^{-2} Gy. On estime que ces concentrations sont trop faibles pour constituer un danger pour la santé, aussi bien celle des animaux que celle des consommateurs.

Mots-clés : Résidus d'usine d'uranium; Ra-226, lièvre d'Amérique; absorption de radionucléides; déchets radioactifs de faible concentration.

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CONTENTS

	Page
ABSTRACT	i
RESUME	ii
INTRODUCTION	1
METHODS	2
RESULTS AND DISCUSSION	3
CONCLUSIONS	12
ACKNOWLEDGEMENTS	12
REFERENCES	12

FIGURE

No.

1. Sampling locations in the Elliot Lake area 3

TABLES

1. Ra-226 content, dose rate and lifetime cumulative dose received by hind leg bones of snowshoe hares, *Lepus americanus* 6

2. Grouped frequency distribution of Ra-226 levels in stomach contents of snowshoe hares, *Lepus americanus*, trapped around uranium tailings and control sites 8

INTRODUCTION

Proper management of the deposits of finely divided wastes (tailings) produced by crushing, grinding and concentrating ore during uranium mining and milling operations requires knowledge of the release rate of radioactive materials and other contaminants from these deposits, and of any consequent environmental impact after abandonment. Such understanding, coming from assessments made on tailings treated in different ways to stabilize them, is desirable to develop waste management strategies, regulations, and cost-effective methods to protect environmental and human interest. The extensive inactive tailings deposits, containing over 150×10^6 t of processed ore covering more than 600 ha, around the uranium mines near Elliot Lake, Ontario, Canada, are of different ages and have received differing treatments after being decommissioned. These tailings are to be returned to nature after rehabilitation and eventually will be areas of wildlife management. This varied set of deposits permits assessments to be made on the efficacy of the various waste stabilization treatments which have been used. We are currently engaged in examination of radionuclides in the biota of the Elliot Lake region.

Previously, we have shown that the meadow vole (*Microtus pennsylvanicus*), a small microtine rodent grazing on tailings vegetation containing appreciable levels of uranium decay series radionuclides accumulates ^{226}Ra in its skeleton (Cloutier et al. 1985, 1986a,b). Faecal pellets of snowshoe hare (*Lepus americanus*) collected near different uranium mine tailings have been shown to contain mean ^{226}Ra contents in their cores ranging from 69 ± 10 to 648 ± 63 $mBq.g^{-1}$ dry weight (Clulow et al. 1986). Radium being an analog of calcium (Eisenbud, 1973), we believed that these animals might also accumulate this radionuclide in their bones after ingesting

it while browsing in the vicinity of the tailings. We report here on ^{226}Ra levels in bones (femur and tibiofibula) and stomach contents of hares trapped near uranium tailings and on control sites, calculate the average internal skeletal radiation dose due to ^{226}Ra, and discuss the problems of calculating the transfer coefficient of the ^{226}Ra from food to hare.

METHODS

Study sites included three inactive uranium mine waste deposits near Elliot Lake, Ontario (46° 23' N, 82° 39' W): the Stanrock tailings, chemically stabilized by addition of lime slurry to the surface, and the Nordic and Lacnor tailings, which have been revegetated (Figure 1). Collection sites were downwind from the wastes and as close as possible to their boundaries. Prevailing winds in excess of 18.5 km/h are from the North and West as measured at the Elliot Lake Airport (Dave et al. 1985). Control sites were located approximately: a) 3 km east of the Nordic tailings (C-1) (Figure 1): b) 15 km NW of the closest uranium tailings in Elliot Lake (C-2) (Figure 1): and c) 880 km east of Elliot Lake at Disraeli, Quebec (45° 54' N, 71° 22' W).

Hares were snared during winter (Jan. to March) where tracks were observed on the snow surface. No attempts were made to snare animals in the absence of snow cover as runways were poorly defined due to the rocky terrain. Hare samples (frozen because of winter temperatures -10 to -30°C) were transported to the laboratory and thawed. Hare size and weight were recorded (Anderson 1965), then they were skinned and dissected. Stomach contents were removed and oven dried at 70°C for one week. Bones of one hind leg (femur and tibiofibula) and both upper arms (humeri) were removed and cleaned by scraping. The proximal ends of both humeri were scoured in commercial bleach solution (Gross and Gross 1966) to reveal more clearly the presence or absence

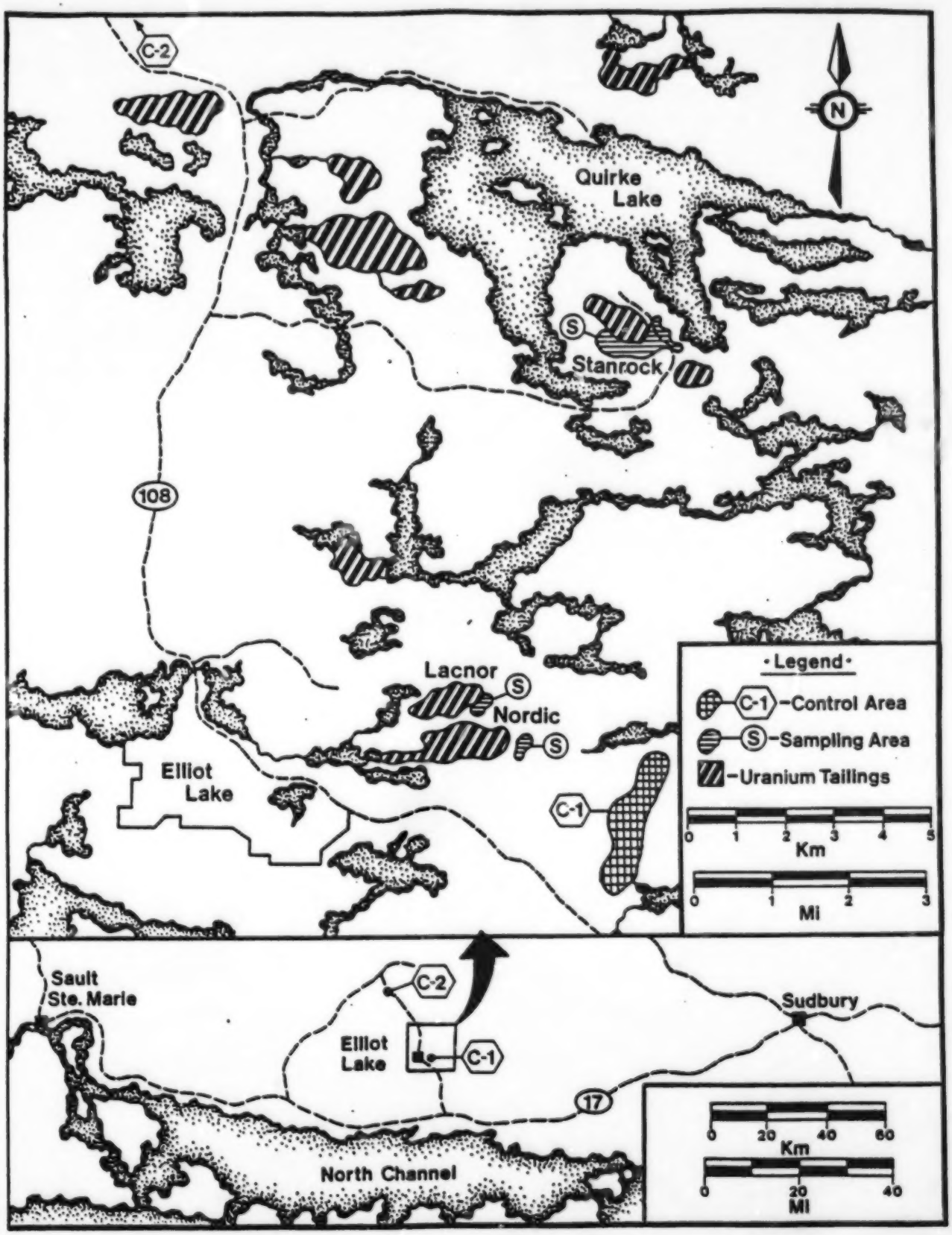

Fig. 1 - Sampling locations in the Elliot Lake area.

of epiphyseal cartilages used in age determinations (Hale 1949; Dodds 1965). Bones of the hind leg were oven dried at 70°C for one week, cut into 2-3 cm lengths, cooled in a desiccator for one hour and weighed. Additional time in the oven did not produce an appreciable weight loss. Moisture loss of the bones when dried to constant weight was 27.3 ± 0.63% (n=9). Bone samples were slowly ashed in a muffle furnace at a maximum temperature of 550°C, for at least 8 h. Ashes were then crushed, sufficient of the oxidant NH_4NO_3 (30%) added to wet them, and oven dried at 70°C overnight. Next day, samples were re-ashed at 550°C for over 12 h then dissolved in 10% HCl, spiked with a measured amount of ^{133}Ba tracer, and diluted to 1 L. An aliquot of the final solution was analyzed for ^{226}Ra content. Tests of the digestion and radio-analytical techniques, using pairs of beef shank bone samples, with or without known amounts of ^{226}Ra and ^{133}Ba added, indicated a ^{226}Ra recovery rate of 98 ± 10%. Stomach contents were digested following the procedure described for faecal pellets in Clulow et al. 1986.

Ra-226 concentrations were measured, following precipitation as Ra-Ba sulphate from solution samples, by alpha-spectroscopy in which the 4.78 MeV alpha-decay peak of ^{226}Ra was counted. Analytic and measuring methods are described in detail elsewhere (Lim and Dave 1981). The ^{133}Ba tracer solution was added to the starting solid sample to measure the overall chemical recovery of ^{226}Ra analysis.

Statistical analyses of data were carried out using the SPSS software package (Nie et al. 1975). Differences among bone ^{226}Ra concentrations at various sites were tested for significance at the 5% level by one way analysis of variance (ANOVA); where significance was indicated in the ANOVA, a Student-Newman-Keuls range test was used to separate sample means different at the 5% level.

RESULTS AND DISCUSSION

As snowshoe hares are born only up to the end of August (Meslow and Keith 1968) and all our animals were trapped in January, or later, they were clearly at least four months old. To distinguish juvenile from adult hares, several methods were tried. Although Keith et al. (1968) identified eye lens weight as the surest indicator of juvenile males after the month of December, and juvenile females after mid-March, this method was unusable because of the breakdown and loss of lens tissue due to the freezing which occurred (unavoidably) in the field (Friend 1967). Hale's (1949) method for aging cottontail rabbits, based on regression of the epiphyseal cartilage of the humerus, has been adapted for snowshoe hare aging by Dodds (1965). All our samples showed a complete absence of the cartilage. Using Dodds' criteria this would indicate an age of 235 days (approximately 8 months) or more. Since Dodds cautions that ossification may vary individually, and among populations in different areas, and that this may cause error in aging wild animals, it is possible that some of our samples may have been younger than eight months. We conclude that bone development was complete in our specimens and that they were certainly at least four months, and probably at least eight months old.

Table 1 presents the levels of ^{226}Ra measured in bones of animals from various sites. Animals taken near Stanrock and Lacnor tailings had levels of ^{226}Ra about ten times higher ($P<0.05$) than those from either control site in the region of Elliot Lake. As only two specimens were taken from the area of the Nordic tailings their data were not included in the statistical analyses, however, these two animals had levels approaching those of the controls. These values are to be compared with the average ^{226}Ra concentration in dried vegetation (211 ± 22 mBq.g^{-1}) and tailings substrate ($11,900 \pm 370$ mBq.g^{-1})

Table 1 - Ra-226 content, dose rate and lifetime cumulative dose received by hind leg bones of snowshoe hares, Lepus americanus

Site	n	^{226}Ra in Leg Bones C_d $mBq.g^{-1}$	Dose Rate dD/dt $Gy.d^{-1}$	Cumulative Lifetime Dose D Gy
Stanrock tailings (chemically stabilized)	6	$220 \pm 32^{a*}$	2.6×10^{-5}	2.8×10^{-2}
Lacnor tailings (revegetated)	4	275 ± 94^{a}	3.2×10^{-5}	3.5×10^{-2}
Nordic tailings (revegetated)	2	20(average)	2.3×10^{-6}	2.6×10^{-3}
Control C-1 (3 km east of Nordic)	6	30 ± 10^{b}	3.5×10^{-6}	3.8×10^{-3}
Control C-2 (15 km NW of U mines)	5	26 ± 7^{b}	3.0×10^{-6}	3.3×10^{-3}
Control C-3 (880 km east of U mines)	5	$<3.7^{**}$	$<4.3 \times 10^{-7}$	$<4.7 \times 10^{-4}$

* Mean ± 1 S.E. indicated, values with different superscripts significantly different at the 5% level (ANOVA/Student-Newman-Keuls range test, 'Nordic' and 'Control C-3' omitted, see text).

** Below detection limit.

(Cloutier et al. 1986b).

Lower levels of ^{226}Ra in faecal samples from Nordic compared to Stanrock and Lacnor tailings, reported previously (Clulow et al. 1986), and bone levels, noted here, may reflect lower levels in the diet of the Nordic animals compared to those of the other tailings sites. At both Stanrock and Lacnor, perhaps due to the abundant forest vegetation which attracts animals and provides them with protective cover, hares approach and penetrate the tailings areas. Signs of hare feeding activity were visible on the snow surface of the Lacnor tailings in winter. On the other hand, the Nordic site lacks forest cover and is surrounded by open country for 200 m; it seems reasonable to suppose that animals from this site do not appoach but browse at some distance from the waste deposit (where there is forest cover) and that vegetation in their diet contains less ^{226}Ra than at the other two sites.

Based on the ^{226}Ra levels in hare faeces reported earlier (Clulow et al. 1986), we concluded that airborne contaminants at Control C-1 were too low to enter into the diet of the animals in appreciable amounts, although the control site is within the downwind range (6.6 km), where at such distances contamination by airborne particulates has been recorded in the U.S.A. (Skinner 1982). Data from the present study support our previous conclusions: ^{226}Ra levels in bones of hares collected at Control C-1, located 3 km downwind of the Nordic tailings were similar to those of Control C-2, located 15 km upwind from the closest uranium tailings.

Ra-226 levels in skeletons of hares collected in an area far removed from Elliot Lake (Control C-3) were all below detection limits (<3.7 $mBq.g^{-1}$ dry weight) and therefore at least five times lower than background levels in the Elliot Lake area (Controls C-1, C-2) (Table 1). Statistical comparison of data from all three control sites was not possible because Control C-3 levels were not quantified. The higher levels of the Elliot Lake control samples

Table 2 - Grouped frequency distribution of Ra-226 levels ($mBq.g^{-1}$ dry wt.) in stomach contents of snowshoe hare, Lepus americanus, trapped around uranium tailings and control sites.

Site	Class Limits ($mBq.g^{-1}$ dry wt.) <3.7	3.8-35.0	35.1-55.0
Stanrock tailings	5	0	1
Lacnor tailings	3	0	1
Nordic tailings	2	0	0
Control C-1	6	0	0
Control C-2	5	0	0
Control C-3	5	0	0

probably reflect a generally higher background level of the radionuclide related to the rich uranium deposits in the region (Dave et al. 1985).

In Table 2 is seen the distribution of ^{226}Ra levels in stomach contents at the time of capture. These values were quite different from the faecal levels reported previously (Clulow et al. 1986). With the exception of one sample from Stanrock and one from Lacnor, all samples analyzed contained ^{226}Ra levels lower than the detection limit of 3.7 $mBq.g^{-1}$ dry weight and considerably below levels in faeces. Although considered to have a sedentary nature Lepus americanus has a home range area which may extend to several hectares (Boutin 1979) and animals are known to make temporary but long-range movements from these centres of activity on occasion (Boutin et al. 1985). These observations carry the implication that feeding and defaecation of the hares may occur in widely spaced locations. Consequently, faeces and stomach contents of animals collected on or near tailings do not necessarily contain forage taken in the vicinity of the high ^{226}Ra levels. Further, the levels in faeces may reflect higher body excretion rates affected by dietary variations in $^{226}Ra/Ca$ ratio and total body burdens. As the measured levels are so heterogeneous, it is not possible to calculate a transfer coefficient from food to animal, based on the small stomach content data as obtained.

Hares established near uranium tailings are exposed to such external radiation sources as radon gas and its decay products, by inhalation, and internal ones such as ingested radionuclides, which may accumulate in the body. Although calculation of the dose from external sources is beyond the scope of our study, our knowledge of ^{226}Ra levels in the skeleton permits a calculation of the radiation dose from Ra decay series alpha-particles, which have major biological effects. Beta-ray and gamma-ray contributions have been omitted from the dose calculation (after Van Dilla et al. 1958; Harley and Pasternack 1976; Momeni et al. 1976).

According to Momeni et al. (1976), the time-integrated dose rate, i.e., the cumulative dose, based on the energy from alpha-particles, plus recoil energy produced by each ^{226}Ra disintegration as 4.86 MeV, and ^{222}Rn and its radioactive daughters as 20.36 MeV, are:

for ^{226}Ra:

$$D_{Ra} = 1.38 \times 10^{-8} \times 4.86 \int_{to}^{t} C(t)dt \qquad \text{Eq 1}$$

and for ^{222}Rn and its daughter products:

$$D_{Rn\ daugh.} = 1.38 \times 10^{-8} \times 20.36 \int_{to}^{t} C(t)f(t)dt \qquad \text{Eq 2}$$

where, C(t) is the average skeletal radium concentration in fresh bone ($mBq.g^{-1}$) at time t (days), and f(t) is the radon-to-radium ratio in the skeleton. The unit of dose is the Gray (Gy).

The radon:radium fraction, f(t) in the skeleton is assumed to be the same as the total body radon retention. Harley and Pasternack (1976) suggested alpha-dose computations from ^{226}Ra based on a radon:radium ratio of 1:3. In a previous paper (Cloutier et al. 1986a) the total dose from ^{226}Ra and its daughter products to the bone was thus obtained by combining Equations 1 and 2, using a radon:radium ratio of 1:3, and by expressing C(t) in terms of measured ^{226}Ra based on dry weight C_d ($mBq.g^{-1}$ dry weight), and moisture content fraction 'm' to give:

$$D = 1.6 \times 10^{-7} \times C_d \times (1-m) \times t_{max} \qquad \text{Eq 3}$$

where, t_{max} is the age of the animal at the time of capture.

The moisture content 'm' was found to be 27.3% (see methods), and the average dose rate (Gy/day) is:

$$\frac{dD}{dt} = 1.60 \times 10^{-7} \times C_d \times (1-m) \qquad \text{Eq 4}$$

Equations 3 and 4 are based on the assumption that the skeletal level of ^{226}Ra measured was constant throughout the life of the hares. The

assumption leads most certainly to an overestimate of dose calculation as the level of ^{226}Ra was likely to be less before ossification was complete.

Using Equation 4, and the measured skeletal ^{226}Ra concentrations and moisture content, the average dose rate dD/dt for hares near uranium tailings and control sites have been calculated and are shown in Table 1. Because the bones of all specimens showed signs of reaching maximum development, and because the Ra:Ca ratio in the diet is taken to remain constant throughout life, it is assumed that the level of ^{226}Ra determined in the specimens would have been constant in animals for most of their lifetime. In a population of hares on Manitoulin Island, 40 km or so South of the experimental sites, only 3 to 17% of animals were three or more years old in a 1959 study (Newson and De Vos 1964). Considering the published annual adult survival rates (see Meslow and Keith 1968), which range from 0.03 to 0.64 but lie mostly between 0.2 and 0.3, it seems doubtful that many hares live beyond three years in the wild. Consequently, the lifetime cumulative dose was based on the 3-year span and was calculated using Equation 4 and t_{max} = 1095 days.

These calculated doses for snowshoe hare are below the threshold dose rates and lifetime cumulative doses of 3.9×10^{-5} $Gy.d^{-1}$ and 0.8 Gy (man), 1.1×10^{-4} $Gy.d^{-1}$, and 0.5 Gy (dog), and 1.6×10^{-3} $Gy.d^{-1}$, and 1.1 Gy (mouse) required to produce osteosarcoma (Raabe et al. 1983). It seems unlikely therefore that the ^{226}Ra will adversely affect the hares of the area. As skeletal tissue is less easily assimilated by predators and scavengers the spread of ^{226}Ra through trophic levels is probably quite slow.

As ^{226}Ra may not distribute evenly in the skeleton, some areas (hot-spots) may in fact have a higher ^{226}Ra concentration than measured and therefore be subjected to a higher radiation dose (Momeni et al. 1976).

CONCLUSIONS

Snowshoe hares living in the immediate vicinity of revegetated uranium tailings have elevated amounts of ^{226}Ra in their skeletons. This finding reflects the situation in meadow voles (Microtus pennsylvanicus) established on the same tailings (Cloutier et al. 1985). The calculated lifetime cumulative doses of 2.6×10^{-3} to 3.5×10^{-2} Gy (based on the average ^{226}Ra levels in the hind leg) are believed to be too low to produce osteosarcoma in animals of the area. The measured levels of ^{226}Ra in the skeletal tissues (20-275 $mBq.g^{-1}$ d.w.) are too small, compared to levels in the tailings, to contribute significantly to the environmental transport of radioactivity from the tailings to secondary consumers, including man.

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